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DEPARTMENT OF THE AIR FORCE HEADQUARTES, 603D REGIONAL SUPPORT GROUP (USAEF)

FINAL REPORT

The first period of the project, due 31 December 1997

Title of Project:

Supersonic Chemical Oxygen-Iodine Laser Driven by

Jet Singlet Oxygen Generator

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I. INTRODUCTION

A development and investigation of chemical oxygen-iodine lasers is aimed to two main application programs - a program of industrial laser technologies with the interest in material cutting, drilling, hardening (e.g., in a ship industry or a nuclear power stations technology), and a program of military laser applications with defence interests (e.g., Airborn laser program under the USEF).

Reasons of concern about COIL utilization in various fields are obvious and could be summarized as follows:

- ♦ an availability and a relative cheapness of chemicals used in production of species for the pumping process of COIL,
- ♦ high specific energy (>150 J/g) released from chemicals providing a high storage of energy in the laser active medium which results in
 - a high overall efficiency of the laser system
 - a high output power on a kilowatts level
- a wavelength of 1.315 μm suitable for
 - a very efficient transmission of laser beam by accessible optical fibres
 - a high absorption by metal surfaces of processed materials
 - a good propagation through the atmosphere
 - an use of non-linear optical technique
- a low pressure of active medium results in
 - a good homogeneity of active medium in resonator
 - a good beam quality (a small beam divergence, a good beam focusability)
- a possibility to operate the laser in CW and pulsed regime
- ♦ a theoretical presumption of scaling-up the device to megawatts power level which predetermines COIL for special technologies

An improvement of operation parameters and increase in the chemical efficiency of powerful facilities require a **fundamental and promotional research** on particular problems (both theoretical and experimental) performed **on a small-scale devices.**

A COIL operates on the electronic transition between the excited and ground states of atomic iodine, $I(^2P_{1/2})\rightarrow I(^2P_{3/2}) + h\nu$ (1.315 µm). The upper laser level is pumped by a near resonant energy transfer from the first excited state of molecular oxygen (singlet delta state), $O_2(^1\Delta)$ to iodine atom in a ground state

$$O_2(^1\Delta) + I(^2P_{3/2}) \leftrightarrow O_2(^3\Sigma_g) + I(^2P_{1/2})$$
 (1)

In current investigations, a **supersonic regime of COIL** is preferred that is achieved by an isentropic expansion of gas in the gain region. The advantages of supersonic operation can be summarized as follows:

- ♦ The equilibrium of pumping reaction (1) is shifted substantially to the right owing to a temperature dependence of equilibrium constant (K_{eq} =0.75 exp (402/T)). A threshold concentration of $O_2(^1\Delta_g)$ is substantially decreased for getting a positive laser gain. Owing to this, a higher proportion of singlet delta energy can be extracted as a laser power.
- ♦ A laser gain is enhanced owing to a high mass-flow rate.
- ♦ Density gradients degrading a laser beam quality are reduced due to a stream stretching wise gain region .
- ♦ A size of COIL device is scaled down substantially at the same output power.

A process of $O_2(^1\Delta)$ generation by a gas-liquid chemical reaction according to a stoichiometry

$$Cl_2(g) + H_2O_2(l) + 2KOH \rightarrow O_2(^1\Delta) + 2H_2O + 2KCl$$
 (2)

must take place in a very thin boundary layer (of the order of 10^{-8} m) near a gas-liquid interface because of extremely short lifetime of $O_2(^1\Delta_g)$ ($\sim 2~\mu s$) in liquid. In this way, the loss of $O_2(^1\Delta_g)$ by quenching in liquid phase is minimized.

A demand of supersonic COIL for $O_2(^1\Delta_g)$ of a high pressure of several kPa and yields $([O_2(^1\Delta_g)]/[O_2]_{total} > 0.5)$ has been satisfied by two types of singlet oxygen generator: a rotating disk SOG and a **jet SOG**.

Characterization of jet SOG

- It is a very effective high pressure source of $O_2(^1\Delta_g)$ up to 13 kPa (100 Torr).
- ♦ A large and controlled specific surface area of gas-liquid interface guarantees a high mass transfer rate.
- ullet A large reaction surface allows to reduce a reaction volume, and, owing to a high gas velocity, also to reduce $O_2(^1\Delta_g)$ loss in gas phase.
- A high jet velocity through reaction zone ensures a fast renovation of boundary liquid layer and a low $O_2(^1\Delta_g)$ loss in liquid phase.
- ♦ An efficient heat transfer eliminates a gas heating and limits a production of detrimental water vapour.
- An increase in jets velocity to 20 30 m/s can provide O_2 up to 100 Torr with more than 50 % of $O_2(^1\Delta_g)$. The energy carried by $O_2(^1\Delta_g)$ thus corresponds to ~ 770 W from the 1 cm² of cross-section of jet SOG which is the highest value of all types of generators used in COIL so far.
- ♦ A gas-liquid counter-flown system ensures good conditions for a self-cleaning of gas flow from liquid droplets which allows to operate the COIL without a droplets separator.
- ♦ A generator possesses no moving mechanical.
- ♦ A possibility of scaling-up the generator to very high specific surface area (cm⁻¹) exists.

Brief history of COIL investigation in the Institute of Physics, Academy of Sciences of the Czech Republic in Prague

An experimental and theoretical investigation of COIL related problems started in 1985 by study of processes in low pressure singlet oxygen generator (bubbler SOG)¹⁻⁴ used to

operate a subsonic COIL with the output power up to 200 W^{5,6}. The operation characteristics of this laser device as a small signal gain, a saturation intensity, etc., were measured.⁷ Further, a pulsed regime of laser operation was investigated. It utilized a gain modulation by externally exposed magnetic field directly to a resonator active region.⁸⁻¹⁴ Some procedures from this period are protected by the Czech and the USA patents.

Since 1996, the work in COIL laboratory has been concentrated mostly to the experimental investigation of jet-type SOG for a supersonic COIL. A designed and constructed small scale jet SOG was used to study the operation characteristics and the output parameters (i.e., $O_2(^1\Delta_g)$ yield, residual chlorine and water vapour concentrations) that were interpreted by a mechanism of processes taking place in both the liquid and gas phase during $O_2(^1\Delta_g)$ production. A great attention was paid to evaluation of water vapour content in gas flowing from a jet SOG to a laser active zone because of a detrimental effect of water on lasing. The experimental conditions (e.g., jets and gas velocity though a generator cavity, peroxide and hydroxide concentration in the generator liquid (BHP solution), a liquid temperature, a total pressure in the generator cavity, dimensions, shape and arrangements of liquid jets, etc.) were determined to suppress a content of water vapour to minimum corresponding approximately to the saturated water vapour pressure. $^{15-19}$

In addition to this main activity, a theoretical study of mixing problems in COIL has been performed by its modelling in the form of 1D kinetic model. A part of working capacity was devoted to the investigation of $O_2(^1\Delta_g)$ generation by a photochemical method and to the utilization of chemically generated $O_2(^1\Delta_g)$ in a fullerene reactivity study 22,23.

An overview of important results was presented at the "Window-on-science,, seminars organized by the USAF Phillips Lab at Kirtland AF Base, NM, and the USAF Office for Research and Development at Bolling AF Base, Wash DC. The talk titled "COIL related activities in the Czech Republic, was given by J. Kodymová (1996), the talk "Singlet oxygen generator for a supersonic chemical oxygen-iodine laser. Parametric study and recovery of chemicals, was given by O. Špalek (1997).

II. PROJECT CHARACTERIZATION

A project was opened on the 1st September 1996 and has to do with investigations in a supersonic Chemical Oxygen-Iodine Laser (hereafter abbreviated as COIL) driven by a jet singlet oxygen generator (SOG). An originally designed and constructed device has to fulfil the following requirements:

- ♦ to be as compact and simple as possible despite its operation complexity,
- ♦ to operate the laser at a relatively high value of pressure in a subsonic region (~70 Torr),
- ♦ to achieve a stable laser output power when a liquid recirculation in the jet SOG is employed.

A construction of the jet SOG and the investigation of generator parameters have to be performed with an upgraded vacuum pumping complex to the point where it will handle a supersonic COIL. A design and a construction of the optical diagnostic chamber have to be made to determine a concentration of singlet delta and sigma states $(O_2(^l\Delta_g), O_2(^l\Sigma_g))$ of oxygen, a residual chlorine concentration, and a water vapour content in gas flowing to a laser duct. A design and a construction of the iodine management, the gases (Cl_2, He) management, the supersonic nozzle, and the laser resonator have to be made. An output coupling measurements, a gain distribution beyond the mixing zone, an energy extraction efficiency, and other related physical parameters of supersonic COIL with the output power up to 1 kW have to be investigated.

Finally, physical characteristics of the supersonic COIL driven by the jet SOG should be compared with characteristics measured with the COIL driven by the rotating discs SOG (VERTICOIL) developed at the Air force Research Laboratory/Directed Energy.

A fabrication, an installation, and a testing the individual parts of the COIL device have to proceed according to the project schedule.

III. PROJECT SCHEDULE

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IV. DESCRIPTION OF ORIGINAL DESIGN OF SUPERSONIC COIL DRIVEN BY JET SINGLET OXYGEN GENERATOR

1. Device subsystems - General drawing # 365 (Supplement Page I)

A supersonic Chemical Oxygen-Iodine Laser (COIL) driven by a jet SOG was designed to have the output power comparable approximately with the output power of VERTICOIL system developed in the USAF Phillips Laboratory driven by a rotating disks generator. A design was made for a molar flow rate of chlorine up to 70 mmol s⁻¹. Dimensions of the device were derived from a pumping power of 3000 m³ h⁻¹ of the upgraded vacuum pumping complex RUTA (Leybold AG). It corresponds to about 75 % of the engine pumping power used with VERTICOIL.

A jet singlet oxygen generator (JSOG) (item 3000) has a cross-section of 30 cm² (50 x 60 mm) and a jet length of 10 cm or 15 cm, respectively. It is designed in a counter-flowing arrangement. Jets are driven by a difference between the outlet pressure of a gear pump used for liquid circulation and the operating pressure in the jet SOG (30 - 100 Torr).

For technical details see description of Assembly drawing 365-3000 (Supplement Page II).

A BHP tank (item 3300) was designed for a preparation of BHP (basic hydrogen peroxide) solution by mixing 70 % H₂O₂ and 50 % KOH, and a BHP maintenance at defined temperature before and during a jetting. It has a cylindrical jacket and a volume of 30 1 approximately. The liquid inside the tank is cooled by a pipe cooler made of stainless steel (316 L). Ethylalcohol used as a coolant circulates between this cooler and a commercial (Czech Comp. HHK Corp.) ribbed heat exchanger - 1 made of stainless steel (the input power of 1.8 kW, a cooling power of 2,3 kW at -30 °C). A centrifugal pump - 2 (Grundfos' Comp.) of the pumping power of 30 l min⁻¹ and able to operate at low temperatures (~ -25 °C) is employed for alcohol circulation. A shut-off electropneumatic valve - 7 is used to evacuate the tank through a gas line parallel to the gas path used during lasing (see *Assembly drawing 365-3300*, Supplement Page III).

A gear pump - 3 (Liquidflo's Comp., model 312; pumping rate variable up to 100 l min⁻¹; wetted parts made of stainless steel 316, PEEK and Teflon) is used for BHP pumping from the tank to the generator. During mixing of a precooled H₂O₂ (to -50°C) and a precooled KOH, a parallel pumping line is utilized which leads from the bottom of tank to its top (bypassing the generator). The lines made of polypropylene tubes are used.

A gaseous chlorine is introduced into the generator from a chlorine pressure vessel through a reduction valve, a flow meter and an electromagnetic valve - 4 (see General drawing 365, Supplement Page I). A primary He is admixed to chlorine before entering the oxygen generator. A secondary He flow is introduced into an iodine reservoir as a carrier gas of molecular iodine vapours, and also for blowing over inner mirrors of resonator. An inert gas management consists of a pressure gas vessel, a reduction valve, a flow meter, and an electromagnetic valve 8.

A shut-off elektropneumatic gate valve - 5 (UHV-VAT, Leybold AG) is placed between the generator exit and the detection cell. The gate valve made of stainless steel 316 L is used for shutting-off the gas flow from the generator. A rectangular cross-section of gas channel in the valve is 50 mm x 9.5 mm. A detection cell (item 3106) is used for diagnostics of $O_2(^1\Delta_g)$, $O_2(^1\Sigma_g)$ and residual Cl_2 (Assembly drawing 365-3100).

An iodine management (item 3400) consists of an iodine reservoir (vaporizer) in the form of heated quartz tube, an optical cell for iodine concentration measurement, and iodine injector – 3104. Iodine system is designed for I₂ molar flow rate up to 1.4 mmol s⁻¹. A molar flow rate is evaluated from the spectral absorption of I₂ at 530 nm, a flow rate, pressure, temperature in the optical cell (the iodine injector - Assembly drawing 365-3100, the iodine vaporizer and the optical cell - Assembly drawing 365-3400).

A gas mixture $(O_2 + I_2 + He)$ flows to a **supersonic nozzle** (item **3100**) in a single horizontal slit configuration. A design of the nozzle is made for the mixture of $O_2 + He$ (1:4) $(\kappa = 1.564)$, $h_{crit} = 6.7$ mm (a critical nozzle height), and the Mach number equal to 2.

Report as the examples only. The other Assemble drawings, as well as Detail drawings, of the rest described parts of COIL device were included in Report 003 served on EOARD on 30 May 1997.

Jet SOG - Assembly drawing 365-3000 (Supplement Page II)

A jet SOG consists of pneumatic valve - 1, 10, 22 for opening and closing a BHP supply into reaction volume of the generator, a jet injector in the form of orifice plate made of polyamide with holes drilled in a certain arrangement, a reaction space of the cross-section 50 x 60 mm and the total height of 220 mm. A vertical distance between chlorine supply tubes and outlet gas channel is 100 mm or 150 mm, respectively. A generator body is made of a Plexiglas plates screwed together and sealed by a silicone adhesive. Stainless steel shanks of diameter 6 mm are used to joint the cover - 2, the liquid volume body - 3, 5, 7, the intermediate frame - 9, the jet injector - 11, and the reaction cavity body - 4, 6, 8. Pure chlorine or chlorine-helium mixture, respectively, is supplied into the generator by stainless steel (316 L) supply tubes - 14 of diameter 10/8 mm having two rows of openings of a diameter 1 mm. The first row contains 11 openings, the second row 12 openings in a distance of 5 mm. Jet injectors (orifice plates) have 513 of 0.8 mm orifices or 684 of 0.6 mm orifices. respectively, corresponding to a specific surface area of jets of 4.3 cm⁻¹. Near the outlet gas channel, the orifice plate contains two rows of thin stainless steel tubes (instead of holes) which overlap a channel mouth in order to reduce a liquid escaping. A constriction of the outlet gas channel is performed by a hand-operated throttle valve (choke) - 17, 18. The gas exits the generator through a flange - 12 into a gas channel of a cross-section 50 mm x 9.5 mm. A liquid supply into the generator is through a flange - 13. A tube - 16 is used to measure the pressure in the upper part of reaction cavity. A thermocouple - 21 placed in the bottom part of one reaction cavity wall is used to measure a jet temperature. The generator is attached to the cover of BHP tank.

BHP tank - Assembly drawing 365-3300 (Supplement Page III)

BHP tank consists of a cylindrical jacket - 1, a cover - 2 and a bottom - 3, a cooler

welded from stainless steel parts - 16-28, a stirrer - 13, 14, a tube - 5 for liquid outlet, and a thermocouple well - 11 placed in the tank bottom. The jacket is made of HDPE, the cover and the bottom are made of Plexiglas. A polypropylene flange - 6 is fixed to the bottom for clamping the outlet polypropylene tube. A stainless steel (316 L) socket - 7 in the cover is used for the tank evacuation, and a similar socket - 4 is used for the liquid supply (H₂O₂, KOH) or BHP circulation by the gear pump. The entrance tube - 25 and the exit tube - 26 of the cooler go through the tank cover, as well as the tube for a gas pressure measuring - 9.

Diagnostics cell - Assembly drawing 365-3100

A diagnostics cell - 6 made of polypropylene includes the main gas channel of the cross-section 50 x 9.5 mm leading from the generator to the supersonic nozzle. Further, it comprises a perpendicular small channel in which a Germanium photodiode is clamped by the flange -11 for measuring a fundamental emission of $O_2(^1\Delta_g)$ at the wavelength of 1270 nm. In the opposite direction, there is another gas channel with a Silicon photodiode (clamped by the flange - 10) used for measuring of $O_2(^1\Sigma_g)$ fundamental emission at 760 nm. A channel for measuring a residual chlorine concentration (from the light absorption at 330 nm) goes perpendicularly in the vertical direction through a centre of flange - 12 and further through a window - 20 to a photometer sensor.

Iodine management - Assembly drawing 365-3400

The iodine management consists of the quartz tube - 1, the valves - 2, 3, the optical cell - 4, and the iodine injector - 5 A quartz tube of i.d. 25 mm and 1 m long heated by a heating band is used as the iodine reservoir and evaporator. According to estimation of heat transfer rate, this iodine source is sufficient for a designed laser. A mixture of iodine vapour and helium flows from the evaporator through the heated tube and the valve - 2 into the optical cell for the measuring an iodine concentration by a photometry method at the wavelength of 530 nm. Upstream the cell, an iodine vapour can be diluted by helium (controlled by the valve - 3). Iodine vapour flows from the optical cell into the iodine injector by heated PTFE tubes - 6.

Iodine injector - Assembly drawing 365-3100

Iodine injector 3104 consists of four stainless steel (316 L) quadrangular prisms brazed by silver. A slot of 10 x 10 mm is milled out in the horizontal prisms, on which a 2 mm thick plate is welded. It contains two rows of drilled openings. In the first row (in the axis of the slot) there are 21 openings of i.d. 0.8 mm, and in the second row there are 42 openings of i.d. 0.4. An iodine vapour is supplied through two tubes of diameter 10/8 mm from above and bottom of the injector body. Two power transistors are attached to the injector body for its heating, its temperature is measured by a thermocouple - 25.

Supersonic nozzle - Assembly drawing 365-3100 (Supplement Page IV, Page V)

The supersonic nozzle body is constructed of Plexiglas plates screwed together and sealed by a silicon adhesive. An inner channel of the body has a cross-section of 65 mm x 30 mm, into which a nozzle walls are inserted. A critical cross-section of the nozzle is 50 x 6.7 mm. A calculated profiles of nozzle horizontal walls are additionally opened by an angle of 3°. Flat vertical walls of the nozzle are also opened by 3°. A tube for measuring a static pressure inside the supersonic volume goes through the upper wall of the nozzle body and the nozzle wall, respectively. A stagnation pressure is measured by a Pitot tube (see *Cut D-D*, *position* 14) fixed in the side wall of nozzle body. **Distance plates - 3** with flanges are fixed to the side walls of nozzle body for clamping the arms of optical resonator. The spacers (distance pieces) of 200 mm long are placed between the flanges and the optical arms. The diagnostic cell and the nozzle body are joined by angles - 5 and screws - 28.

Intermediate body - Assembly drawing 365-3200

The intermediate body made of Plexiglas plates is placed between the nozzle body and the liquid nitrogen trap of entrance rectangular cross-section of $400 \times 35 \text{ mm}$.

V. DESIGN OF SUPERSONIC NOZZLE

CALCULATIONS FOR SLIT SUPERSONIC NOZZLE DESIGN

1. Calculation of the adiabatic constant of gas mixture, κ

$$\frac{1}{\kappa - 1} = \sum_{i} \frac{x_i}{\kappa_i - 1} \tag{1}$$

where x_i is a molar fraction of i-gas.

This relation was used to calculate adiabatic constants of gas mixtures considered for use in supersonic Chemical Oxygen-Iodine Laser (COIL).

Tab. 1. Adiabatic constants calculated for gas mixtures for COIL

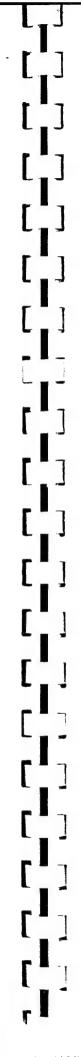
Gas	N ₂	O ₂	Не	O ₂ +He (1:4)	O_2+N_2 (1:4)	O_2+N_2 (1:2)
κ	1.401	1.398	1.630	1.564	1.400	1.399

2. Calculation of gas velocity in the critical cross-section of nozzle

From the Bernoulli equation for adiabatic flow, Saint Venant's and Wantzel's relation follows in the form

$$v_{\text{erit}} = \sqrt{\frac{2\kappa}{\kappa - 1} \cdot \frac{p_{\theta}}{\rho_{\theta}} \left[1 - \left(\frac{p_{\text{erit}}}{p_{\theta}} \right)^{\frac{\kappa - 1}{\kappa}} \right]}$$
 (2)

where p_0 and ρ_0 are primary gas pressure and density, respectively (upstream the nozzle), and p_{crit} and ρ_{crit} are gas pressure and density, respectively, in the nozzle throat.



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As there is a critical gas velocity in the nozzle throat, $v_{crit} = a_T$ (sonic speed at temperature T)

$$a_{\rm T} = \sqrt{\kappa \frac{p_{\rm crit}}{\rho_{\rm crit}}} \tag{3}$$

the following relation can be derived to calculate a critical pressure ratio:

$$\frac{p_{\text{crit}}}{p_0} = \left(\frac{2}{\kappa + 1}\right)^{\frac{\kappa}{\kappa - 1}} \tag{4}$$

By substitution (4) into (2), v_{crit} can be calculated by

$$v_{\text{crit}} = \sqrt{\frac{2\kappa}{\kappa + 1} \cdot \frac{p_0}{\rho_0}} \tag{5}$$

The ratio $P_{\text{o}}/\rho_{\text{o}}$ can be expressed by the gas equation

$$\frac{p_0}{\rho_0} = \frac{R T}{\overline{M}} \tag{6}$$

where M is the mean molecular weight of gas mixture

$$\overline{\mathbf{M}} = \sum_{\mathbf{i}} \mathbf{x}_{\mathbf{i}} \mathbf{M}_{\mathbf{i}} \tag{7}$$

Critical velocity

a) for the gas mixture of O_2 +He (1:4): M = 0.0096 kg, $\kappa = 1.564$

The ratio P_0/ρ_0 amounted to 227 770 m² s-² (T = 298 K) and the critical velocity in nozzle throat according to (5) gave

$$\mathbf{v}_{\text{crit}} = 561 \text{ m s}^{-1}$$

b) for gas mixture of O_2+N_2 (1:2): M = 0.0288 kg, $\kappa = 1.399$

The calculations gave the ratio P_0/ρ_0 equaled to 86027 m² s⁻² (T = 298 K) and

$$v_{crit} = 316.8 \text{ m s}^{-1}$$

3. Calculation of dimensions of nozzle channel

a) for the gas mixture of O2+He (1:4)

Input parameters:

$$v_{crit} = 561 \text{ m s}^{-1}$$
 $n_{Cl2} = 100 \text{ mmol s}^{-1}$
 $n_{He} = 400 \text{ mmol s}^{-1}$
 $n_{\Sigma} = 0.5 \text{ mol s}^{-1}$
 $T_o = 263 \text{ K}$ (temperature upstream the nozzle)
 $Q_r = 0.83 \text{ m}^3 \text{ s}^{-1} = 3000 \text{ m}^3 \text{ h}^{-1} \text{ (Roots pump capacity)}$
 $P_o = 6.6 \text{ kPa (50 torr)}$
 $w = 50 \text{ mm}$ (nozzle width)
 $T_r = 298 \text{ K}$ (temperature upstream the pump)

The flow rate in the nozzle critical cross-section amounted to

$$\dot{V}_{crit} = \frac{\dot{n} R T_0}{P_0} = 0.188 \text{ m}^3 \text{ s}^{-1}$$

Then the nozzle cross-section:

$$S_{crit} = \frac{\dot{V}_{crit}}{V_{Activ}} = 3.35 \text{ x} 10^{-4} \text{ m}^2 = 3.35 \text{ cm}^2$$

and the critical nozzle height:

$$h_{crit} = \frac{S_{crit}}{W_{crit}} = 0.67 \text{ cm} = 6.7 \text{ mm}$$

Pressure at the upstream the pumping system:

$$P_r = \frac{\dot{n} R T}{Q_r} = 1486 Pa = 11.1 torr$$

b) for the gas mixture of O_2+N_2 (1:2)

Input parameters:

 $v_{crit} = 317 \text{ m s}^{-1}$ $n_{Cl2} = 100 \text{ mmol s}^{-1}$ $n_{N2} = 200 \text{ mmol s}^{-1}$

 $n\Sigma = 0.3 \text{ mol s}^{-1}$

 $T_o = 263 \text{ K}$ (temperature upstream the nozzle) $Q_r = 0.83 \text{ m}^3 \text{ s}^{-1} = 3000 \text{ m}^3 \text{ h}^{-1}$ (Roots pump capacity)

 $P_0 = 3500 \text{ Pa} = 26 \text{ torr}$

 $V_o = 0.187 \text{ m}^3 \text{ s}^{-1}$

w = 50 mm (nozzle width)

 $T_r = 298 \text{ K}$ (temperature upstream the pump)

The flow rate in the nozzle critical cross-section amounted to

$$\dot{V}_{crit} = \frac{\dot{n} R T_0}{P_0} = 0.187 \text{ m}^3 \text{ s}^{-1}$$

Then the nozzle cross-section:

$$S_{crit} = \frac{\dot{V}_{crit}}{V_{crit}} = 5.9 \text{ x} 10^{-4} \text{ m}^2 = 5.9 \text{ cm}^2$$

and the critical nozzle height:

$$h_{crk} = \frac{S_{crit}}{W_{crk}} = 1.18 \text{ cm} = 11.8 \text{ mm}$$

Pressure at the upstream the pumping system:

$$P_r = \frac{\dot{n} R T}{Q_r} = 892 Pa = 6.7 torr$$

4. Calculation of supersonic nozzle shape

A method of characteristics^{24,25} was used to calculate the supersonic nozzle shape for its design. The calculation was performed for gas mixtures considered for experiments, i.e., a mixture of O_2 -He (1:4), and O_2 - N_2 (1:2) with adiabatic constant $\kappa = 1.564$ and 1.399, respectively. The Mach number 2.0 in the flow at the end of the nozzle, a number of the characteristics n=40, and a number of the streamlines m=9 were considered.

a) Gas mixture of O_2 +He (1:4)

In **Tab. 2** (Suplement Page IX), calculated values of normalized coordinates of the upper nozzle wall (X,Y), coordinates of the upper nozzle wall $(x,y_0, [mm])$, and coordinates of the lower nozzle wall $(x, -y_0, [mm])$ for $\kappa = 1.564$ and $h_{crit} = 6.7$ mm are given. A normalized length unit is equal to half-height of the nozzle throat $(h_0/2)$. The 7^{th} and 8^{th} columns give coordinates of upper and lower nozzle wall opened by $\pm 2^o$ against the theoretical values. This broadening is made in order to compensate an influence of boundary layers. The 9^{th} and 10^{th} columns give coordinates of nozzle walls opened by $\pm 4^o$. The vertical cross-section of the calculated nozzle shape and its parameters are shown in **Fig. 1** (scale 4:1) (Supplement Page X). The values of the Mach number, M, and thermodynamics data $(P/P_0, T/T_0, \text{ and } \rho/\rho_0)$ at the exit from the nozzle were also evaluated in these calculations. They amounted to M = 2.0, $P/P_0 = 0.123$, $T/T_0 = 0.469$, and $\rho/\rho_0 = 0.261$.

b) Gas mixture of $O_2 + N_2$ (1:2)

In **Tab. 3** Supplement Page XI), calculated values of normalized coordinates of the upper nozzle wall (X,Y), coordinates of the upper nozzle wall (x,y_o, [mm]), and coordinates of the lower nozzle wall (x, -y_o, [mm]) for $\kappa = 1.399$ and $h_{crit} = 11.8$ mm are given. The vertical cross-section of the calculated nozzle shape and its parameters are shown in **Fig. 2** (scale 4:1) (Supplement Page XII). The value of the Mach number and thermodynamics data at the nozzle exit amounted to M = 2.0, $P/P_o = 0.127$, $T/T_o = 0.555$, $\rho/\rho_o = 0.230$.

The angle of 2° was chosen for the nozzle walls opening. After ending a calculated nozzle shape, the ceiling and bottom of the duct are flat forming angels + 2° and - 2° with the plane of the nozzle symmetry. The side walls of supersonic nozzle form the angle \pm 2° with a vertical plane of symmetry in the part extending to downstream to resonator arms and further the walls are parallel. The distance between the supersonic nozzle throat and the optical axis of the resonator is 55 mm, and can be shortened easily for 35 mm.

VI. SUMMARY OF DEVICE SUBSYSTEMS PARAMETERS

1. Jet SOG

Cross-section: $50 \times 60 \text{ mm}$; total length ~ 200 mm; inner volume ~ 600 cm^3 ;

length of reaction zone 10 and 15 cm;

Jet injector : orifice plates: orifice of 0.8 mm, # 513, $S = 4.3 \text{ cm}^{-1}$; $v_{0.8} \sim 93 \text{ l min}^{-1} = 1.57 \text{ l s}^{-1}$

orifice of 0.6 mm, # 684, $S = 4.3 \text{ cm}^{-1}$; $v_{0.6} \sim 70 \text{ l min}^{-1} = 1.16 \text{ l s}^{-1}$

Jets driving force: pressure difference between the atmospheric pressure in BHP tank and

pressure in JSOG, i.e., ~ 0 mbar; $v_{liq} = 600$ cm s⁻¹;

Cl₂ supply: via commercial water chlorinator equipped with flow meter or orifice (sound

flow);

Cl₂ injector: 2 tubes made of stainless steel 9 / 6.5 mm, each 23 holes of 1 mm

Cl₂ molar flow rate: 50 - 100 mmol s⁻¹; counterflowing regime;

He primary flow rate: $\sim 200 - 400 \text{ mmol s}^{-1}$ (Cl₂: He $\sim 1 : 4$); admixed to Cl₂ flow

Total pressure in JSOG: 30 - 100 torr

BHP: 70 % H_2O_2 , 50 % KOH, $[HO_2] \sim 7.8 \text{ M}$; $t_{BHP \text{ input}} \sim -20 \text{ to } -30 \text{ }^{\circ}\text{C}$

Initially: BHP flow down system

Later: BHP flow closed loop

2. BHP Reservoir (mixing tank)

BHP mixing tank: inside a cooling stainless steel coils of cryostat; a stirrer in centre;

a thermocouple monitoring a mixture temperature;

70 % H₂O₂ precooled to - 50 °C, addition a precooled 50 % KOH;

BHP reservoir: total volume ~30 l, 26 cm inner diameter, ~50 cm high,

material: high density polyethylene,

stainless steel tube spiral coil inside with spirit flowing from heat exchanger

Heat exchanger: input power 8 - 10 kW (10 kJ s⁻¹); cooling power \sim 2 kW;

made by Czech Company HKK Corp.;

running temperature of -20 °C to -30 °C;

BHP gear pump: Liquidflo's Equipment Co. Inc. (USA company), model 311, pumping rate 75 l min⁻¹, max difference pressure 5 bar; wetted parts made of 316 stainless steal and HDPE

3. Subsonic channel

Choking valve downstream JSOG: *Bellows sealed gate valve,* Leybold AG, Germany, material: stainless steel, Viton, electropneumatically operated, switching time 0.8 s

Pressure downstream the valve: $\sim 20 - 70$ torr

Gas mixture: He + O_2 (4:1); $u_{crit} \sim 561 \text{ m s}^{-1} (\sim 1.53 \text{ x } 330 \text{ m s}^{-1})$

Diagnostic cell: detection of $O_2(\Delta_g)$, $O_2(^1\Sigma_g)$, residual Cl_2 ;

pressure: gauge CM1000, Capacitron DM 22, Leybold AG, Germany;

4. Iodine injector and vaporiser

I 2 injector: 2 injectors, each with two rows of holes aligned perpendicular to flow:

the first row 21 holes of 0.8 mm, the second row 42 hoes of 0.4 mm;

material : stainless steal, heated by a semiconductor heater (250 W) to \sim + 80 ° - 100 ° C;

distance between I2 injection and critical nozzle plane: 9 and 5 mm;

I₂ detection: molar flow rate evaluated from spectral absorption at 488 nm, He flow rate,

pressure and temperature in the optical cell;

 I_2 vaporizer : quartz tube filled with crystalline I_2 , He as a carrier gas flowing through I_2

crystals layer;

 I_2 molar flow rate: 0.5 - 1.3 mmol s⁻¹

He secondary flow rate: 30 mmol s⁻¹

 $I_2 / O_2 \sim 0.01 - 0.02$

5. Supersonic nozzle for mixture O_2 + He (1:4)

A single horizontal slit configuration

 $\kappa = 1.564$

 $n_{Cl2} = 100 \text{ mmol s}^{-1}$

 $n_{He} = 400 \text{ mmol s}^{-1}$

 $n\Sigma = 0.5 \text{ mol s}^{-1}$

 $T_0 = 263 \text{ K}$ (temperature upstream the nozzle)

 $Qr = 0.83 \text{ m}^3 \text{ s}^{-1} = 3000 \text{ m}^3 \text{s}^{-1}$ (Roots pump capacity)

 $P_0 = 6.6 \text{ kPa} (50 \text{ Torr})$

w = 50 mm (nozzle width)

 $T_r = 298 \text{ K} \text{ (temperature upstream the pump)}$

 $v_{crit} = 561 \text{ m s}^{-1}$

 $V_{crit} = 0.188 \text{ m}^3 \text{ s}^{-1}$ (flow rate in critical nozzle cross section)

 $S_{crit} = 3.35 \text{ cm}^2 \text{ (nozzle cross-section)}$

 $h_{crit} = 6.7 \text{ mm}$ (critical nozzle height)

6. Resonator

Distance of optical axis of resonator from the critical nozzle plane : variable; 35 to 50 mm

Gain length: 5 cm

Resonator arms: 40 cm, made of stainless steal, easily withdrawal, mirrors internally mounted, protected by purging with inert gas;

mirrors 50 mm in diam, active diameter 37 mm, radius 2.63 m, (Czech optical factory Meopta); glass substrate (optical glass BK-7), 10 mm, 9 - 21 dielectric layers

Resonator length: 85 cm

Stable resonator: reflectivity of back mirror: 99.95 %

reflectivity of output mirror: 99.1; 98.9; 98.1; 97.8 % at 1.315 μm (see diagram)

Resonator active area: 37 mm x ~16mm

Aligning and adjusting optical resonator and beam optics (splitter): He-Ne laser LA

001, 50 mW, $\Lambda = 633$ nm;

7. Vacuum pumping system

Roots blower and single-stage rotary pump: RUTA type 3001/2, Leybold AG Germany, $3000 \text{ m}^3 \text{ h}^{-1}$ (0.84 m³ s⁻¹); rotary pump SOGEVAC 630, 630 m³ h⁻¹, vacuum pressure < 3 x 10⁻² mbar; Roots pump RUVAC RA 3001.

Vacuum pipe line: 150 mm in diameter, length 12 m approx.

VII. PROJECT SCHEDULE FULFILMENT IN 1997

Till the end of 1997, the following tasks were fulfilled:

- 1. Upgrading the vacuum complex (purchasing and installation of the new vacuum complex, joining the complex by a vacuum line to a laser device),
- 2. Designing, fabrication and installation of the jet SOG (including a jet orifices plate, a chlorine injector, joining the SOG to a BHP tank, equipping the SOG with a closing elektropneumatic valve and a hand choking valve)
- 3. Design, fabrication and installation of BHP tank (including a thermal management, BHP recirculation by a gear pump, BHP mixer)
- 4. Designing, fabrication and installation of the optical diagnostic cell
- 5. Handwork, design, fabrication and partial installation of the laser (including an iodine injector, a supersonic nozzle, optical arms, mirrors holders, mirrors protecting)
- 6. Fabrication and installation of a diffuser down stream the laser duct
- 7. Installation of liquid nitrogen trap
- 8. Designing and starting the construction of data acquisition by PC on-line (32 channels)
- 9. Designing and starting with fabrication of gas (Cl₂, He) management and I₂ management

An overall view on the constructed supersonic COIL device is presented on the 4 enclosed pictures (Pages VII and VIII in Supplement)

Simultaneously with the fabrication of individual parts of the supersonic COIL, the experiments dealing with the exact calibration of the optical method for determination of $O_2(^1\Delta_g)$ concentration have been performed using the EPR spectroscopy. A small scale device including a chemical generator of bubbler-type has been constructed for $O_2(^1\Delta_g)$

generation and equipped with the optical detection of $O_2(^1\Delta_g)$ and $O_2(^1\Sigma_g)$, respectively, and for $O_2(^1\Delta_g)$ and $O_2(^1\Sigma_g)$ concentration measurement by their electron spin resonance. All the date were collected by PC on-line. The results of this study that have a fundamental significance are processed and prepared for a publication presently.

VIII. COST OF SUPERSONIC COIL DEVICE IN 1996-97

Equipment and Instruments purchased

Vacuum pumping system: Roots blower and single-stage rotary pum	n·RIITA Levhold AG
Germany ¹	\$ 36 000
Heat exchanger : Czech company HKK Ltd.	\$ 7 000
Chock valve, Leybold AG	\$ 1 000
Gear pump, Verder Ltd. Prague (trade representative) ¹	\$ 9 000
Pressure gauges (Capacitron, Leybold)	\$ 1000
Temperature gauges	\$ 1000
Computer data acquisition	\$ 200
Computer data acquisition	\$ 2000
Others	
Constructional material	\$ 2 000
Chemicals, gases	\$ 500
Overhead charges -	\$ 1000
Labour	\$ 6 000
Travel expenses	\$ 2 000
TOTAL:	\$ 67 700
From EOARD support	\$ 35 000
From Local Grant Agencies	\$ 32 700

¹ Paid from local grants budged

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